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Structure and depolarization properties in the Raman spectra of superfluid ^{4}He

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Raman spectra of superfluid ^{4}He under saturated vapor pressure are measured at 1.37 K. Distinct changes in the slope dI_R/dE are confirmed at several energy values in the plot of Raman intensity (I_R) as a function of the energy shift (E) for a measurement with relatively high instrumental resolution. Depolarization ratios are determined up to the energy shift at 420 K. Existence of s partial-wave scattering is confirmed.

I. INTRODUCTION

Halley proposed theoretically that Raman scattering is a useful tool for studying the elementary excitations in liquid helium.[1,2] The first experimental observation of the Raman spectrum of superfluid ^{4}He was done by Greytak and Yan.[3] Since then, measurements of the Raman scattering from liquid helium have been extended to the observation of high-resolution spectra of superfluid ^{4}He,[4–6] of the depolarization ratio of the two-roton peak of superfluid ^{4}He,[7] of the depolarized scattering from the normal state of ^{4}He,[8] of structures in the spectrum of superfluid ^{4}He,[9] of the spectra of ^{3}He-^{4}He mixtures in the superfluid state,[10,11] and of the spectra of condensed states of liquid helium.[12–14] These activities show that Raman scattering is a tool having wide applicability to various states of quantum fluids. Despite those extended measurements, however, there is still no satisfactory theory with which we can quantitatively calculate a spectral shape that closely fits the experimental results over the whole energy region. A number of theoretical works have been done.[2,15–30] The Raman scattering rate at frequency ω is given by[2,15,18,21,22,24,27,28]

$$I(\omega)=K\int\frac{dk}{\rho(2\pi)^3}\int\frac{dk'}{\rho(2\pi)^3}t(k)t(k')H(k,k',\omega)\ ,$$

where K is a constant, ρ is the number density, and k is the wave vector. $t(k)$ is called the light-helium coupling constant. $H(k,k',\omega)$ is called the dynamic correlation function and is the Fourier transform of a four-body correlation function of the density fluctuations. One reason why the theories of Raman scattering are not reliable is that the functional form of $t(k)$ is not well known. Another reason is that the relation between the dynamic correlation function $H(k,k',\omega)$ and the dynamics of the elementary excitations is also not well established. To deduce information about the elementary excitations from $H(k,k',\omega)$, we need to know the relation between the density fluctuation and the boson operators of elementary excitations. A simple way is to assume linear relations between them. We then get four-body correlation functions of the boson operators from $H(k,k',\omega)$. Calculations of the four-body correlation function of the boson operators were carried out with[16,17,22] and without[15,18,20,21] taking into account interactions between elementary excitations.[15,18,20,21] The former calculations gave a good fit with the experimental results at least very near the two-roton region.[4–6] Despite the apparently good agreement, however, the theoretical model used was considered doubtful from the theoretical considerations[27,28] of the excluded-volume conditions proposed by Kleban.[25] The reliability of the linear relation between the density fluctuation and the boson operators was questioned by Nakajima.[18] Thus, theories of Raman scattering from superfluid helium remain controversial without clear conclusions.

The above-mentioned situation warranted more experimental work. We have made accurate measurements over a wide energy-shift region. Two distinct facts have been confirmed: Structures are observed in the spectrum taken with relatively good instrumental resolution, and s partial-wave scattering exists and becomes dominant with the increase of the energy shift. In Sec. II the experimental method and results are described. In Sec. III discussion is given.

II. EXPERIMENT

A. Method

In the experiment, 5145-A radiation, from a Nippon Electric Corporation Model No. GLG3200 argon laser linearly polarized along the vertical direction, was focused to a narrow beam inside the sample cell. To rotate the direction of the polarization of the laser beam to determine the depolarization ratio, a half-wave plate and a polarizer were used. Light scattered at right angles into the horizontal plane was collected with $f/3.7$ optics. The spectrometer was a Jovin Yvon Model No. Ramanor HG2S double-grating monochromator. The light passing through the exit slit was detected by a cooled Hamamatsu TV Model No. R464 photomultiplier tube. A Princeton Applied Research Model No. 1121 photon counter was used for discrimination of the pulse height. The dark count was below 0.7 photoelectron/sec. The data accumulation and the automatic control of the double monochromator were done with a microcomputer. The sample was filled through a filter packed with crushed copper mesh cooled with 4.2-K liquid helium. The temperature of the sample was measured with a carbon resistor and a Texas Instrument Model No. 145 precision pressure gauge.

B. Spectral shape measurement

The results of the measurement of the Raman spectra of superfluid ^{4}He at 1.37 K under saturated vapor pressure are shown in Fig. 1 for three different values of the instrumental full width at half maximum (FWHM); (a) 1.1 K, (b) 1.9 K, and (c) 3.7 K. In these measurements, the laser power was 0.8 W, the incident beam was polarized along the vertical axis, and the polarizer was not used in the light collection system. The unresolved Brillouin doublet was observed as a single central peak. The monochromator reading of the incident laser energy, the integrated relative intensity of the Brillouin doublet I_B, and the instrumental width were determined from the maximum position, the maximum value, and the FWHM of this central peak. Appropriate subtractions of the dark count and the background were made. The Brillouin intensity I_B was always measured before the measurement of the Raman intensity I_R for use as a reference. The relative intensity I_R/I_B is used as the vertical scale of the figures. This quantity is useful for estimating the cross section of the Raman scattering. In order not to detect spurious variation of the Raman intensity due to fluctuations or drift in the mechanical assembly or in the temperature, the spectra were repeatedly measured several times by scanning the energy region shown in Fig. 1. Comparison of the results taken at different scans revealed that no appreciable spurious variations of the Raman intensity occurred. Thus the averages of the total scans were used as the final data. In order to get sufficiently good statistical accuracy, large numbers of signal counts were accumulated. The total number of counts at the maximum of the spectra are 19 333 (750 sec), 17 948 (160 sec), and 20 072 (100 sec) for plots (a), (b), and (c), respectively. The times in the parentheses are the total measuring times per point.

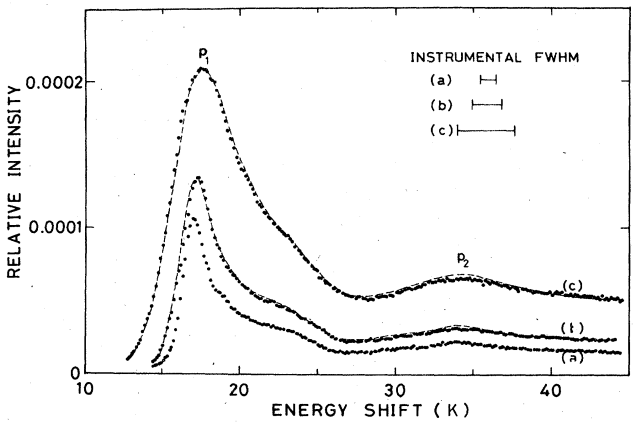

FIG. 1. Raman spectra of ^{4}He at 1.37 K under saturated vapor pressure measured with three different values of the instrumental FWHM; (a) 1.1 K, (b) 1.9 K, and (c) 3.7 K. Dashed curves are results obtained from the convolution of spectrum (a) with Gaussian resolution functions having widths of 0.8 K for plot (b) and 2.6 K for plot (c). These plots show mutual consistency with spectra observed with different instrumental widths.

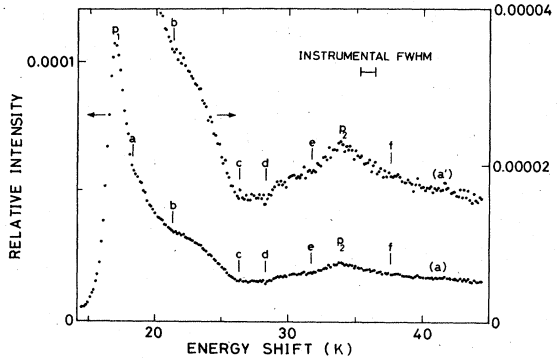

FIG. 2. Raman spectrum of ^{4}He at 1.37 K under saturated vapor pressure measured with the instrumental FWHM of 1.1 K. In the plot (a'), the vertical scale is enlarged three times compared with the plot (a). Changes of the slope are observed at energies labeled as a–f.

structure becomes clear as we reduce the instrumental width down to 1.1 K as shown in plot (a), while spectra (b) and (c), taken with wider instrumental widths, are mostly consistent with those of former works.[3] To show this structure more clearly, curve (a) in Fig. 1 is plotted by expanding the vertical scale as shown in curve (a) in Fig. 2. We can clearly observe several places where the slope dI_R/dE changes. Here E is the energy shift. The positions of those energies are shown with vertical bars labeled as a–f. The structures are most distinct near c and d. To show this more clearly the data are plotted as (a') in Fig. 2, expanding the vertical scale by a factor of 3. Abrupt changes of the slope dI_R/dE are seen at c and d. Between these energies, the Raman intensity is nearly constant. We can note other small changes. Shoulderlike behavior appears in the energy region between a and b, and b and c. We also note small changes of the slope near e and f.

C. Polarization property measurement

To clarify the Raman process, the measurement of the polarization characteristics is important. Former measurements were done in the two-roton energy region.[3,7] We extended our measurements over a wider energy-shift region. The Raman intensity I_R is the sum of the polarized part I_p and the depolarized part $I_{\rm dp}$; $I_R=(\mathbf{e}_i\cdot\mathbf{e}_s)^2I_p+I_{\rm dp}$. Here, $\mathbf{e}_i$ and $\mathbf{e}_s$ are the unit vectors along the directions of the incident beam polarization and the easy axis of the polarizer used in the scattered light collection system, respectively. We write the Raman intensities as I_{vv} observed for the case when both $\mathbf{e}_i$ and $\mathbf{e}_s$ are vertical, and I_{hv} observed for the case $\mathbf{e}_i$ is horizontal and $\mathbf{e}_s$ is vertical; $I_{vv}=I_p+I_{dp}$ and $I_{hv}=I_{dp}$. The results of the measurements of those intensities with 5.4-K instrumental FWHM are shown in Fig. 3. The vertical scale is the relative intensity normalized by the intensity of the central peak observed in the I_{vv} measurement. With repeated experiments, I_{hv}/I_{vv} at the two-roton peak was determined to be 0.74 ± 0.02. This value is consistent with that of Woerner and Greytak.[7] The ratio $I_{hv}/I_{vv}=0.74$ at the two-roton peak was used to normalize and plot I_{hv} in Fig. 3. Between the energy-shift values

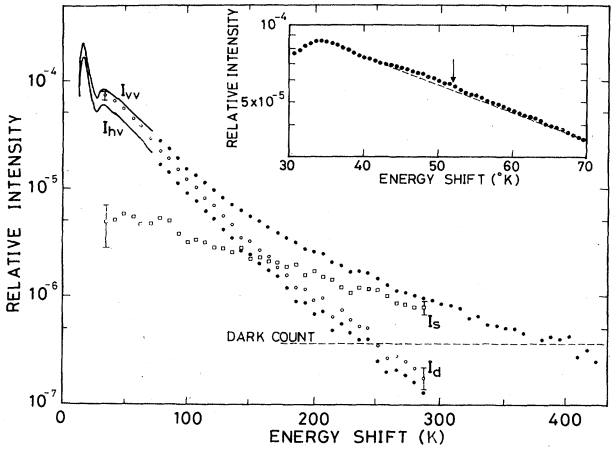

FIG. 3. Raman spectrum of ^{4}He at 1.37 K under saturated vapor pressure measured with 5.4-K instrumental FWHM. For meanings of I_{vv}, I_{hv}, I_s, and I_d, see text. Estimated errors of I_s and I_d are shown at two end points. The horizontal dashed line is the dark count rate of the photomultiplier tube. Inset: I_{vv} near the six-roton energy, shown with an arrow. The straight dashed line is drawn to help the reader see deviations $\log(I_{vv})$ from the linear dependence on energy shift.

of 14.4 and 71.9 K, results are shown with solid curves drawn through the data points because successive data points were taken with a 0.719-K (0.5-cm^{-1}) step overlap and cannot be distinguished. In the energy-shift region above 71.9 K, data are shown with circles taken with a 7.19-K (5-cm^{-1}) step. The experimental results, shown in Fig. 3, show that I_{hv} decreases faster than I_{vv} as the energy shift increases. This means the intensity of the polarized light becomes dominant as the energy shift increases.

Theoretically, the Raman intensity is expected to include s and d partial-wave scatterings. By writing the respective intensities of these scatterings in terms of I_s and I_d, $I_R=(\mathbf{e}_i\cdot\mathbf{e}_s)^2I_s+[\frac{3}{4}+(\mathbf{e}_i\cdot\mathbf{e}_s)^2/4]I_d$, according to Iwamoto.[16] Thus, $I_{vv}=I_s+I_d$ and $I_{hv}=\frac{3}{4}I_d$. Using these relations, I_s and I_d were calculated from the experimental data and the results are shown with open squares and circles in Fig. 3, respectively. The existence of s partial-wave scattering is evident and its contribution to the total scattering becomes dominant as the energy shift increases.

III. DISCUSSION

We make a few brief comments on the facts confirmed in this experiment. The energy shifts and the intensity ratios I_R/I_B of the peaks labeled as p_1 and p_2 in Fig. 1 are listed in Table I. The spectra shown in Fig. 1 were measured without using a polarizer in the detection system, the incident beam being linearly polarized along the vertical direction. The intensity ratios of the peak p_1 were also measured with a polarizer in the detection system with its easy axis along the vertical direction. The results are listed in Table I. The change in the observed energy shift of p_1 with the change of the instrumental resolution has been controversial. Measurements with different instrumental resolutions were done by Greytak *et al.*[3,4] The higher-resolution experiment[4] showed a good fit with the theoretical model of two-roton resonance.[16,17] However, the position of peak p_1 observed in the first experiment[3] with lower resolution (4.2-K instrumental FWHM) did not fit the same theoretical model. This fact was discussed by Woods and Cowley.[22] Hastings and Halley[24] also tried to introduce a localized interaction model to explain the fact, believing both experimental values to be correct. When the instrumental FWHM was increased in our experiment, we could not detect such a large shift in p_1 as observed by Greytak and Yan.[3] As Murray *et al.* have already mentioned, the reproducibility of the energy reading of a grating monochromator is sometimes not good[5] and the first experiment by Greytak and Yan was in error in the energy determination. In fact, in our experiment we needed to observe the position of the central line just before the start of the Raman spectrum measurement and also to scan the spectrometer in the same direction to realize ±0.1-cm^{-1} reproducibility. Our results are consistent with the results obtained with a Fabry-Perot interferometer by Gretyak *et al.*[4,5]

Hastings and Halley calculated the spectral shape over a wide energy-shift region for the case of 4.2-K instrumental FWHM.[24] Although Hastings has mentioned[30] that the calculations are somewhat unreliable due to violation of the excluded volume conditions,[26–28] we will still note one aspect of their calculations. They pointed out the possibility of the appearance of peaks at energies approximately equal to even multiples of the single-roton energy Δ_0, irrespective of the model used. p_1 is observed

TABLE I. Changes of the energy shifts and the intensity ratios I_R/I_B of peaks p_1 and p_2 as the instrumental width was changed. For details see text.

Instrumental width (K)	3.7	1.9	1.1
	Peak p_1		
Energy shift (K)	17.6±0.14	17.3±0.14	17.1±0.07
I_R/I_B (without polarizer)	$(2.09\pm0.05)\times10^{-4}$	$(1.34\pm0.03)\times10^{-4}$	$(1.1\pm0.1)\times10^{-4}$
I_R/I_B (with polarizer)	$(1.88\pm0.03)\times10^{-4}$	$(1.29\pm0.03)\times10^{-4}$	$(0.89\pm0.03)\times10^{-4}$
	Peak p_2		
Energy shift (K)	34.4±0.4	0.34.5±0.4	34.5±0.4
I_R/I_B (without polarizer)	$(0.65\pm0.02)\times10^{-4}$	$(0.33\pm0.04)\times10^{-4}$	$(0.23\pm0.02)\times10^{-4}$

near $2\Delta_0$ as is well known. The observed position of peak p_2 in this experiment is 34.5 ± 0.5 K, which includes $4\Delta_0=34.7\pm0.16$ K, four times the single-roton energy determined by neutron scattering.[31] The six-roton process might be observed near $6\Delta_0$. In the inset of Fig. 3, a measurement of I_{vv} near $6\Delta_0$ with a 5.4-K instrumental FWHM is shown. We note that $\log(I_{vv})$ versus the energy-shift plot is slightly convex upwards near the six-roton energy, denoted with an arrow.

The value of the ratio I_R/I_B has been discussed by several theorists in order to clarify the Raman process. As shown in Table I, this value depends strongly on the instrumental FWHM. We note I_R/I_B changes linearly with the change of the instrumental width. Extrapolations to zero instrumental width give some estimates of the peak height ratios of p_1 and p_2 to the central Brillouin peak. Those values are about 0.5×10^{-4} for p_1 and 0.4×10^{-5} for p_2. We do not compare those values with available theoretical calculations. I_R/I_B was found to depend strongly on temperature in the relatively low temperature range of 1.2—1.5 K. This is partly because I_B is approximately proportional to temperature. But our measurements showed that I_R at p_1 also changed as the temperature was lowered. Most of the theoretical calculations of I_R assume a temperature of absolute zero. Thus, lower-temperature experiments are needed to get a value of I_R/I_B which can be compared with theories.

As shown in Fig. 2, structure becomes clear as we reduce the instrumental FWHM. Ohbayashi and Ikushima once reported that structure exists in the Raman spectrum of superfluid ^{4}He.[9] However, doubts were raised by two groups soon after.[5,14] Since then, it appears to have been believed that there does not exist any structure. The structures observed in this experiment are similar to those observed by Ohbayashi and Ikushima, although they are not exactly the same. Because the spectral shape changes with changes in the experimental parameters such as temperature, we need more detailed, extensive work to clarify fully the relations between various experimental results obtained by different groups under different experimental conditions.

To our knowledge, there has been no quantitative theoretical calculation that has predicted the spectral structures shown in Fig. 2. We denote the energy of the local maximum of the phonon-roton dispersion curve by Δ_1, the roton energy being denoted by Δ_0. Theory relating the Raman spectrum to the state density of the interacting elementary excitation pair[16,17,23] is thought to be basically correct. Hereafter, we denote this density of states by ρ_2. The theory is evidently not completely accurate because of the neglect of the light-helium coupling $t(k)$. However, we can discuss at least the local behavior of the spectrum with ρ_2 if $t(k)$ changes gradually and smoothly. Greytak *et al.*[4] and Murray *et al.*[5] have performed high-resolution measurements of the Raman spectrum near the two-roton energy region. They observed good agreement between the experimental results and ρ_2. The shift of the position of the two-roton peak from $2\Delta_0$ to lower energy was claimed to be evidence for the roton-roton bound state. But their high-resolution measurements were limited in energy to within ±1.5 K of the two-roton energy and cannot claim anything outside this energy region. Our measurements are poorer in resolution compared with theirs, but our measurements were extended over a wider energy-shift region and the resolution was good enough to observe some structure in the spectrum as shown in Fig. 2. As clear structure, we can discuss the behavior near the energies labeled as *c* and *d*. The separation of the two energies is large compared with the instrumental width. We note that the appearance of such distinct structure in the collective mode of a simple fluid may be characteristic of a condensate state of a quantum fluid. The energy labeled as *d* is close to but slightly larger than $2\Delta_1$ as determined by neutron scattering. The local behavior of the Raman spectrum near $2\Delta_1$ is different from the theoretical ρ_2 shown in Figs. 5 and 6 in Ref. 23. According to those figures, ρ_2 has a broad and weak peak below $2\Delta_1$ and decreases sharply when the energy shift approaches $2\Delta_1$ from the smaller energy-shift region. We observed rather a flat region between *c* and *d* as shown in Fig. 2. There is a clear qualitative difference between the present experimental results and ρ_2. Concerning the fact that agreement is good near the two-roton energy as observed by Greytak *et al.*,[4,5] but not good near $2\Delta_1$ as shown in this experiment, we point out the following possibilities. One possibility is that the theory is mostly correct near $2\Delta_0$ but needs some minor modifications, such as local changes of parameters, near $2\Delta_1$. If such is the case, we need not reconsider the roton-roton resonance model. Another possibility may be the discrepancy near $2\Delta_1$ is a manifestation of the incorrectness of the theory. Then we may regard the good fit near $2\Delta_0$ as accidental. We can mention this possibility because Kleban and Hastings proved that the two-roton bound-state model violates the excluded-volume conditions.[27] This is a serious defect. As far as we know, there is no published theory to oppose this proof. According to the theoretical discussions of the excluded volume conditions, we need to take into account the off-diagonal coupling between different momentum states.[27,28] Then even the local spectral shape, including the peak position, is expected to be different from ρ_2. The excluded volume condition is also related to the warning made by Nakajima that the linear expansion of the density fluctuation in terms of boson operators is a crude approximation.[18] If higher-order terms are added, they may contribute to scattering exhibiting spectral shapes different from ρ_2. The discrepancy between our results and the theoretical ρ_2 may be related to those theoretical controversies. To make clear conclusions, however, we need quantitative theoretical calculations of spectral shape that take into account the excluded volume conditions. What we can safely claim from our results is that the discrepancy between the data shown in Fig. 2 and the theoretical ρ_2 clearly shows at least partial incompleteness in the theory in relating the Raman spectrum to the density of states of the interacting quasiparticle pair, ρ_2.

As for the other weak structures (labeled *a*, *b*, *e*, and *f*), they are not so clear, and we need more experiments with improved resolution to clarify them fully. The data shown in Fig. 2 is the best we could do with our present apparatus.

Two aspects of depolarization ratio have been dis-

cussed: the ratio at the two-roton peak and the integrated intensity ratio of s to d partial-wave scattering. Greytak and Yan found that Raman scattered light is highly depolarized.[3] Stephen explained the depolarization using the dipole—induced-dipole model of light-helium coupling,[15] which results only in d partial-wave scattering. Iwamoto pointed out the possibility of both s and d partial-wave scatterings. Nakajima mentioned that not only the dipole-dipole interaction but also the interaction due to the direct overlap of wave functions of near-neighbor atoms is important in two-roton Raman scattering.[18] The latter causes both s and d partial-wave scatterings. Fetter used the convolution approximation to decouple the correlation function and gave a numerical value for the s partial-wave scattering at the two-roton peak.[20] His estimate was $I_{hv}/I_{vv} \simeq 0.69$. Careful measurement of the depolarization ratio at the two-roton peak was done by Woerner and Greytak.[7] They observed $I_{hv}/I_{vv} = 0.74 \pm 0.02$. In the case of d partial-wave scattering without s partial-wave scattering, $I_{hv}/I_{vv} = 0.75$. Thus the result of Woerner and Greytak was not inconsistent with zero s partial-wave scattering at the two-roton peak. Thus the dipole—induced-dipole model was often used to calculate the Raman intensity. The integrated intensity ratio of s to d partial-wave scattering, but not the local depolarization property at the two-roton peak, was calculated by Kleban and Halley semiempirically from experimental uv-absorption data.[26] They found that a considerable amount of s partial-wave integrated intensity can be expected. And considering the fact that the two-roton peak is mostly d, they predicted an increase in s partial-wave scattering in the large energy-shift region. Campbell *et al.* performed a microscopic calculation of the integrated intensity ratio of s to d partial-wave scattering and obtained a range of 0.08—1.88.[29] They considered the excluded volume condition. Kleban and Halley's semiempirical calculation,[26] modified to consider the polarization effect,[29] gave a range of 0.11—1.45. In fact we observed the existence of s partial-wave scattering and its increase in the large energy-shift region as shown in Fig. 3. From the results shown in Fig. 3, the experimental value is estimated to be 0.16 ± 0.03, which lies within the theoretical predictions.

In conclusion, the results obtained in this experiment show that seemingly well-established interpretations of Raman scattering from superfluid ^{4}He are partly incorrect. The local structures of the spectrum near $2\Delta_1$ differ even qualitatively from that expected from the density of states of the interacting elementary-excitation pair, which was used to analyze the spectral shape near the two-roton energy $2\Delta_0$. Until the work of Kleban and Halley,[26] the dipole—induced-dipole model of the photon-helium coupling was used exclusively to estimate the Raman intensity. Existence of s partial-wave scattering confirmed in this experiment shows that this model is incomplete. It is interesting to note that observed integrated intensity ratio of s to d partial-wave scattering is consistent with the theoretical calculation[29] that takes into account the excluded volume condition. There is no theory at present to explain the spectral structures shown in Fig. 2 and the energy dependencies of s and d partial-wave scatterings shown in Fig. 3. If a theoretical model is developed with which we can explain the present results, Raman scattering will become a much more powerful tool for investigating condensate states of quantum fluids.

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